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Ab Initio Determination of Aluminum Conductivity in Warm Dense Matter Regimes*

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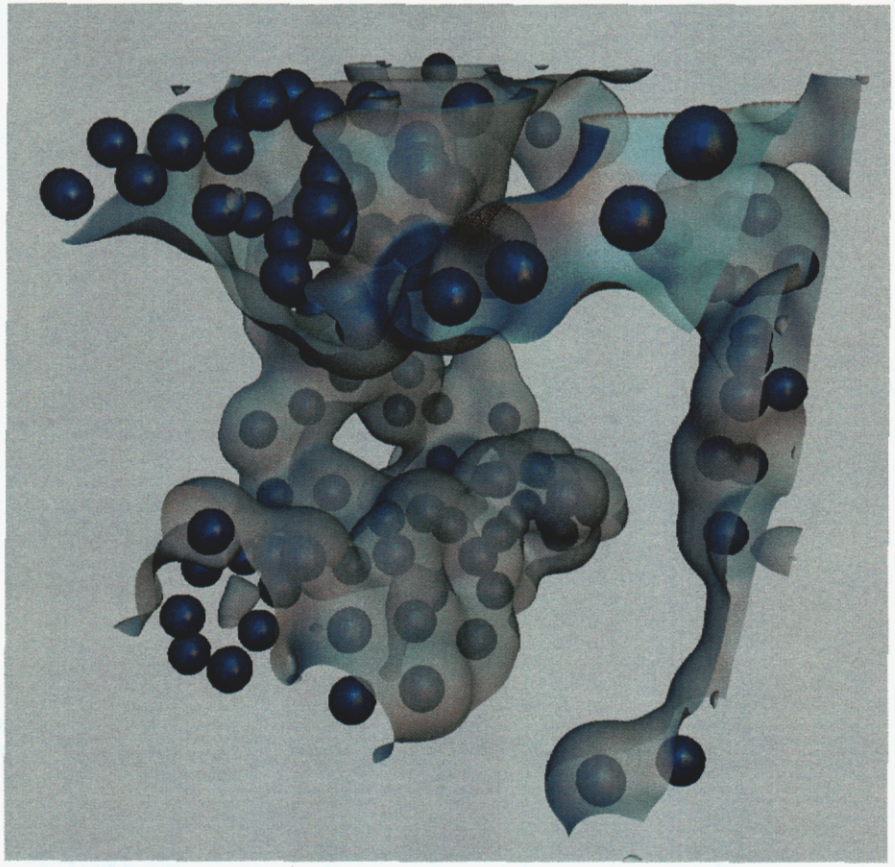
Accurate computer modeling of electrically or magnetically driven high energy density physics experiments in the vicinity of the metal-insulator transition has, in the past, been hampered by large uncertainties in the electrical properties of these materials, and for many materials of interest this is still the case. We have begun a program to improve the accuracy of our conductivity tables and algorithms for these materials --- our most complete treatment to date being that of aluminum. A critical regime of interest for many of the pulsed-power driven experiments at Sandia is from moderate compressions over solid density down to one hundred fold expansions from solid, and temperatures from ambient up to several eV. For much of this region, the electrical conductivity has not been experimentally measured, and analytical results are limited by the uncertainties remaining in modern treatments of these strongly coupled and degenerate, or weakly degenerate systems. To improve our understanding of this warm dense matter regime, we have performed numerous *ab initio* calculations of the electrical conductivity of aluminum using a combination of DFT based molecular dynamics and conductivity calculations with the Kubo-Greenwood formula. Under those conditions for which data is available, the agreement is very good. Using the results from these calculations, in conjunction with data and theory where available and reliable, we have generated a wide range SESAME format table for the conductivity of aluminum. Subsequent computer simulations of high energy density experiments, such as magnetically launched flyer plates and exploding wires, give very good agreement with the experiments.

* Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy under Contract No. DE-AC04-94AL85000.

Ab Initio Determination of Aluminum Conductivity in Warm Dense Matter Regimes



Mike Desjarlais

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High fidelity High Energy Density Physics simulations require good global conductivity and EOS models

Simulations of Z pinches, magnetically insulated transmission lines (MITLs), flyer plates, and exploding wire discharges span several orders of magnitude in density and temperature around the solid state.

Most HEDP codes currently use either the in-line Lee-More algorithm or SESAME tables for the conductivity, principally developed for higher temperature and density regimes.

A particularly complex and important region for these computations is that around the metal-insulator transition.

$$\sigma = n_e A(\frac{\mu}{kT}, \omega\tau)\frac{e^2}{m}\tau$$

Metal-insulator transition, pressure ionization

Degeneracy and magnetization effects

Coulomb and electron-neutral collisions, Bloch-Gruneisen solid, Lindemann melting law

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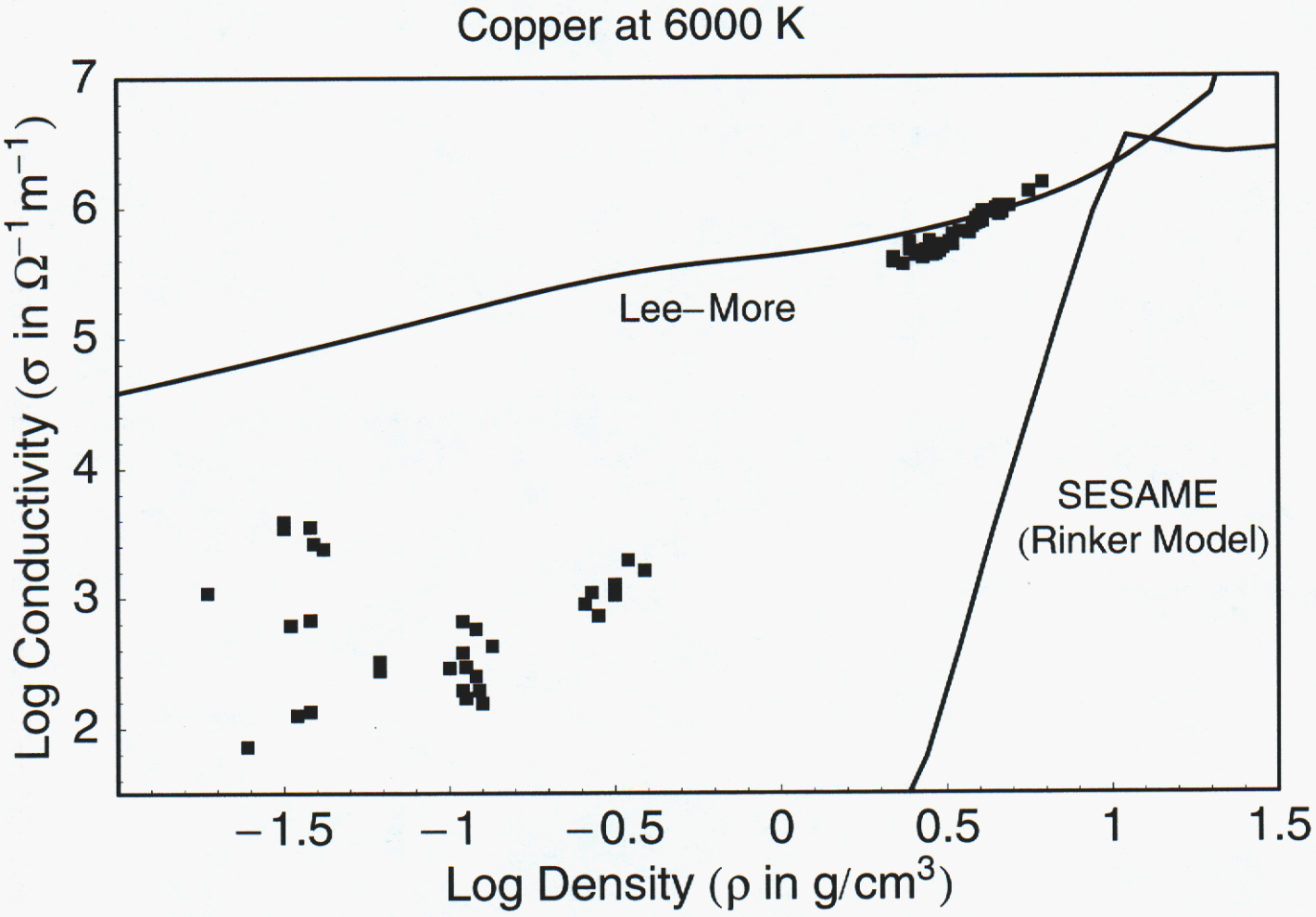
The Lee-More and SESAME (Rinker) models are inaccurate near the metal-insulator transition



Data courtesy of Alan DeSilva, University of Maryland

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Modifications of the Lee-More algorithm have been made to obtain an improved wide-range model*

The most significant changes are:

- **An ionization equilibrium model that blends first-ionization Saha (including pressure ionization) with Thomas-Fermi.**

- **Electron-neutral cross sections are calculated using a screened polarization potential (Born approximation)**

- **The expression for the minimum allowed collision time, $\tau_{\min} = p_2 \frac{R_a}{V_e}$, is generalized to $\tau_{\min} = p_2(n_a) \frac{R_a}{V_e}$, to permit agreement with DeSilva and Katsouros' observations of a density dependent minimum conductivity.**

Other minor changes to the Lee-More parameters are included to allow more accurate agreement with measured solid density conductivities.

* M. P. Desjarlais, Contrib. Plasma Phys. **41** (2001) 2-3, 267-270

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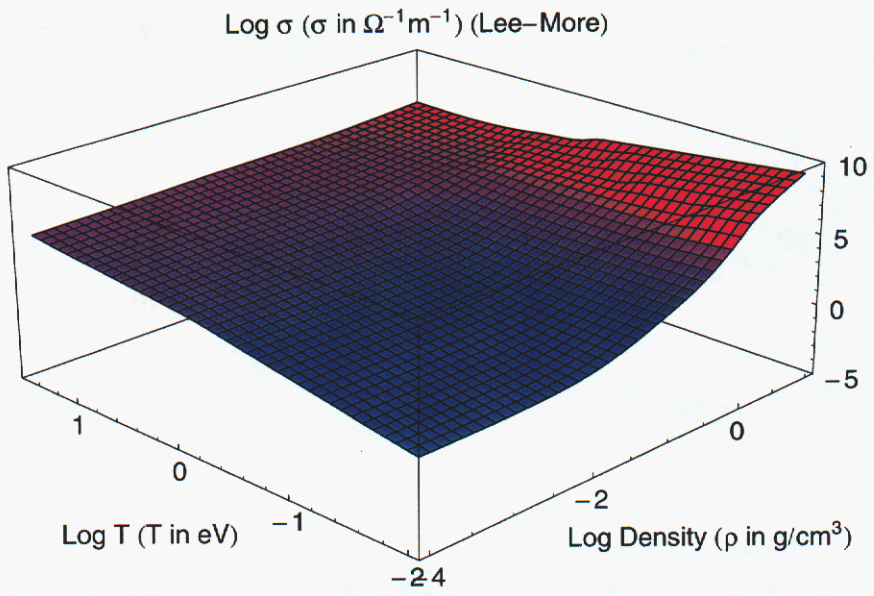
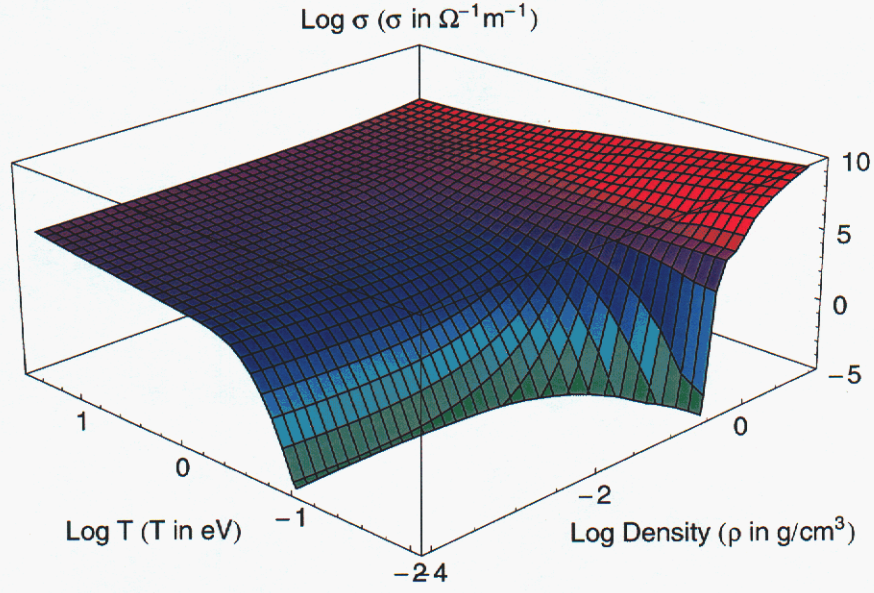
The new model generates a distinct metal-insulator transition





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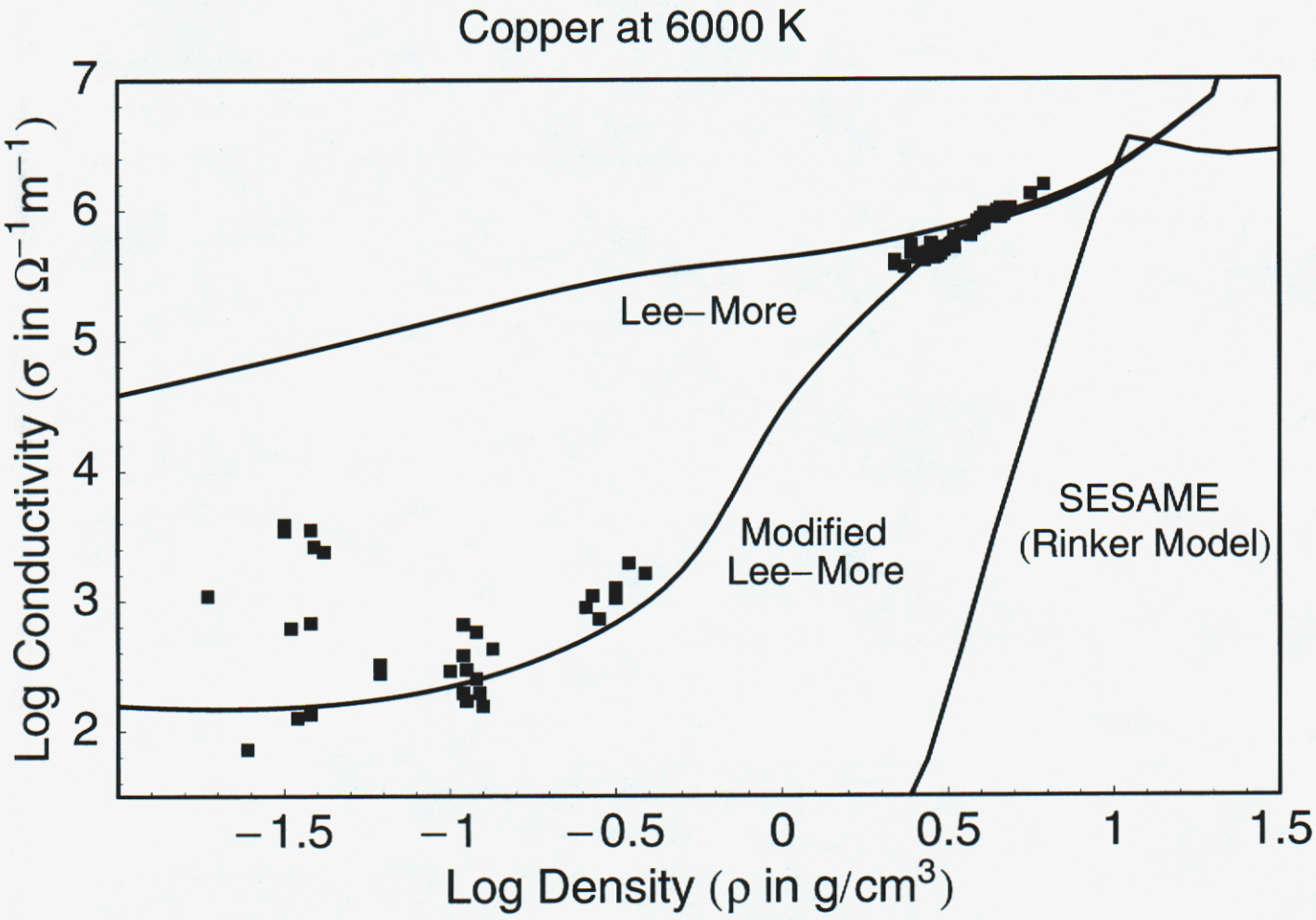
This modified Lee-More model provides much better agreement with DeSilva's copper data



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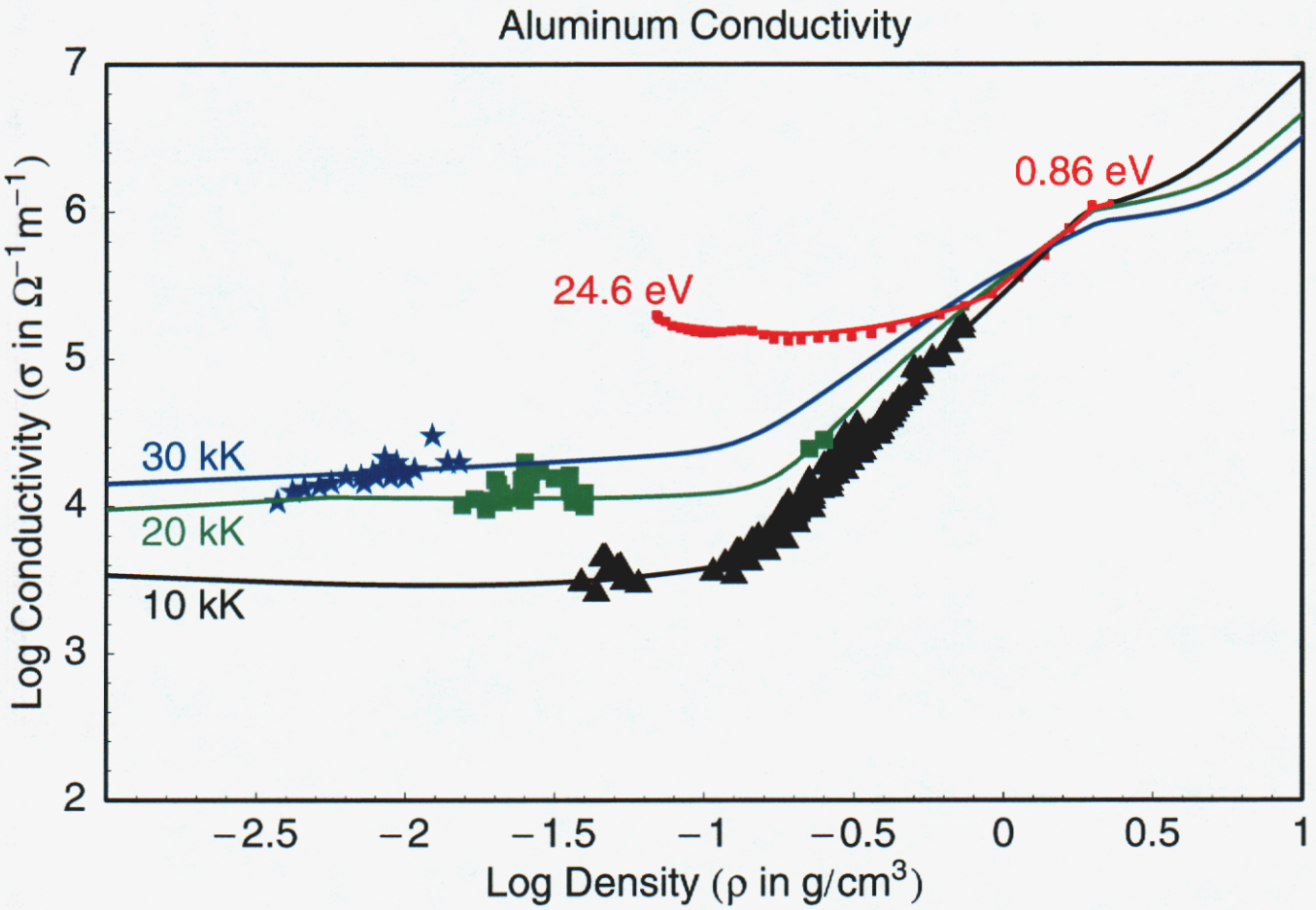
Good agreements with DeSilva's (Maryland) and Benage's (LANL) aluminum data are obtained



The LMD conductivity goes smoothly over to Lee-More at high temperatures and densities

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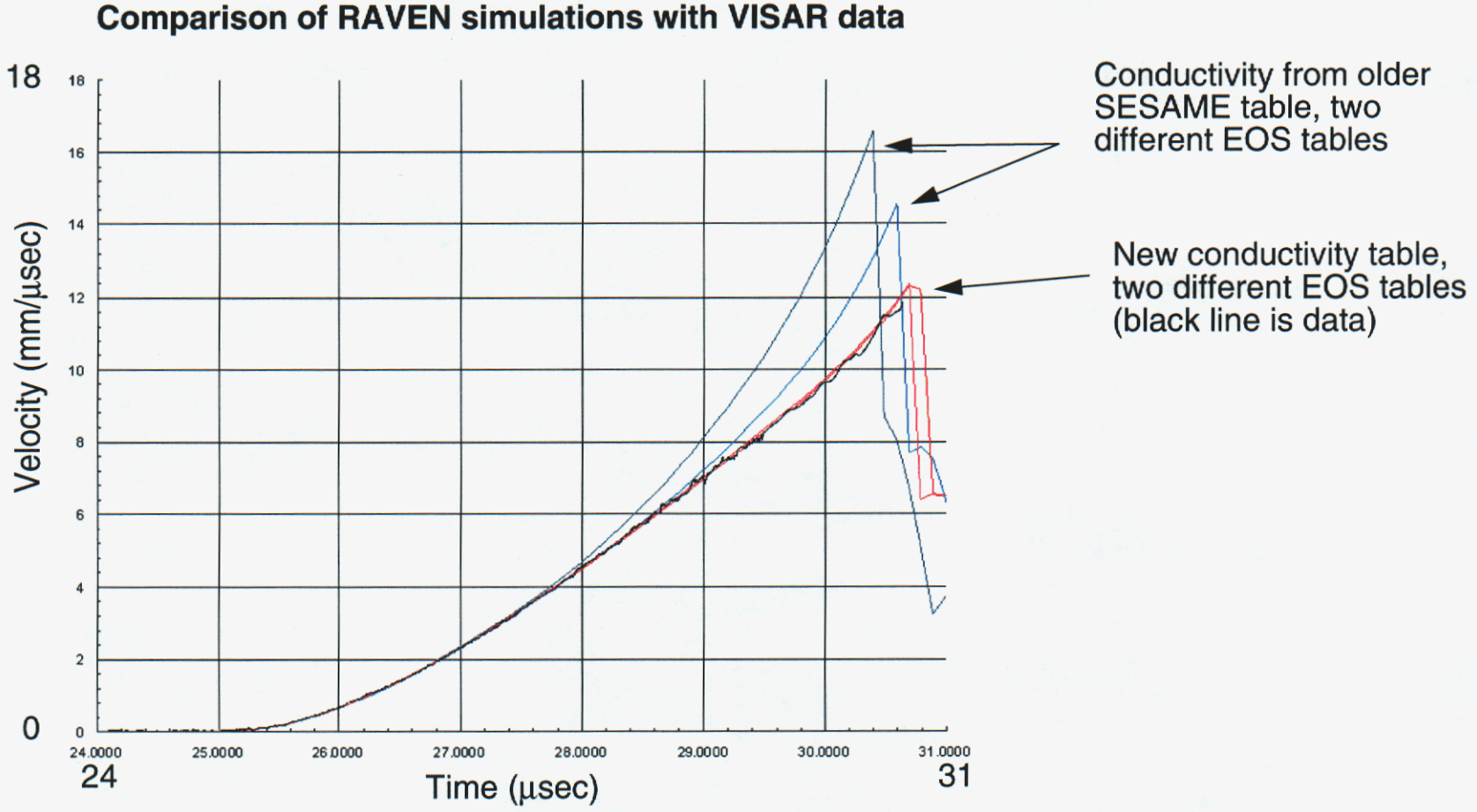
The new conductivities result in better agreement with data for Al liner implosion simulations at LANL



Data and simulation results courtesy of Walt Atchison, LANL

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These improvements were not sufficient for our exploding wire and flyer plate simulations

- Detailed comparison between simulations and experiments for exploding wires (Steve Rosenthal, Sandia) and magnetically launched flyer plates (Ray Lemke, Sandia) suggested that our conductivities were not sufficiently accurate in the dense liquid aluminum regime at moderate temperatures.

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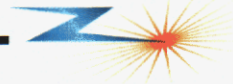
We are using DFT based molecular dynamics simulations to improve our conductivity models



- The MD simulations are performed with VASP (Vienna *Ab initio* Simulation Program), a plane wave density functional code.
- The ions and their respective core wavefunctions are modeled using the Vanderbilt ultra-soft pseudopotentials for the MD runs (very efficient).
- Exchange and Correlation functionals are GGA using Perdew-Wang 91.
- Most of the MD runs use only the Γ point (k = (0,0,0)) to sample the Brillouin zone.
- Simulations use 108 atoms per supercell at the highest densities, and drop to as low as 8 atoms per supercell at the lowest density.
- Typical runs cover one to two picoseconds.

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Following the MD runs conductivities are calculated using the Kubo-Greenwood formula

The Kubo-Greenwood formula provides the optical conductivity directly from the electron wave functions

$$\sigma(\omega) = \frac{2\pi e^2 \hbar^2}{3\omega m^2 \Omega} \sum_{j=1}^{N} \sum_{i=1}^{N} \sum_{\alpha=1}^{3} (F(\varepsilon_i) - F(\varepsilon_j)) \langle \psi_j | \nabla_\alpha | \psi_i \rangle^2 \, \delta(\varepsilon_j - \varepsilon_i - \hbar\omega),$$

where the first two sums are over all N bands in the calculation (typically 300 to 400), the third sum is over the three spatial directions , $F(\varepsilon_i)$ is the Fermi weight corresponding to the energy ε_i for band i, ψ_i is the global wavefunction for band i, and Ω is the super-cell volume for the simulation.

In practice, because of the finite simulation volume, the δ-function must be smeared; we use a Gaussian with a width on the order of the mean eigenvalue separation. We typically average over 10 to 20 configurations, sample over the time history.

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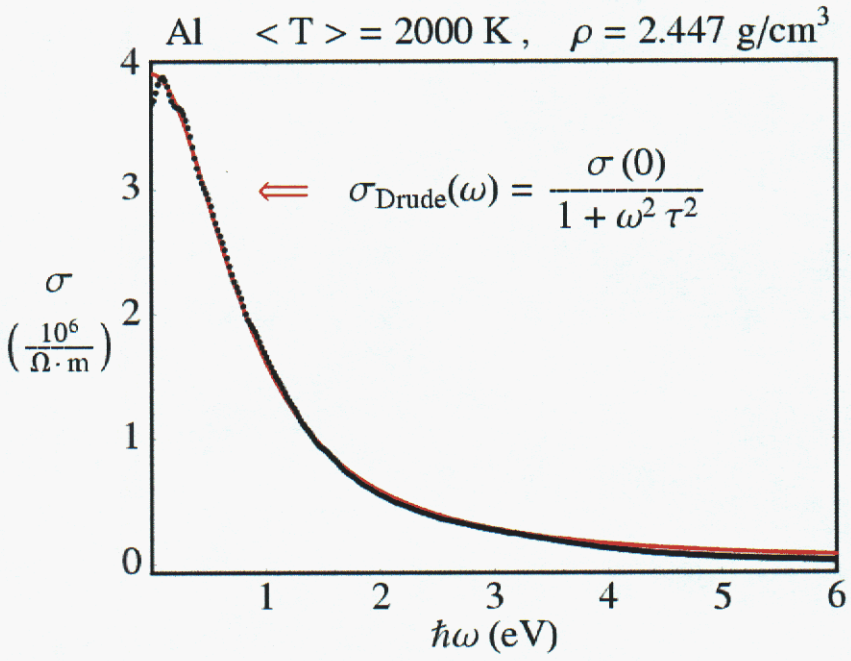
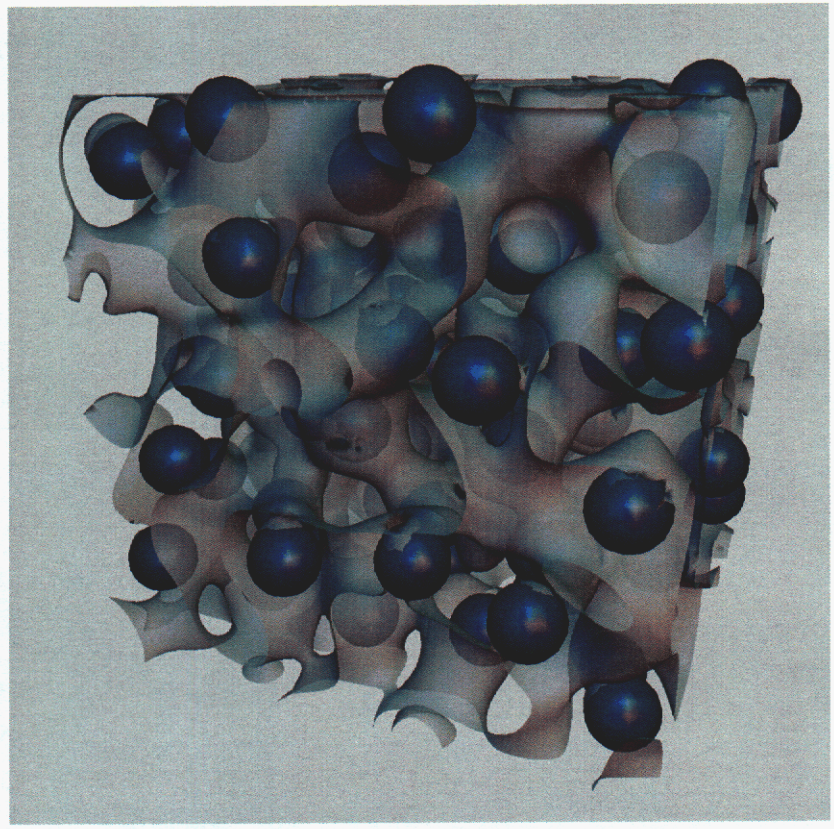
At liquid densities just below solid, the optical conductivity is well fit by the Drude model



The agreement with the Drude model indicates 'nearly free' electrons



Ion cores displayed with iso-surfaces of the mean valence charge density

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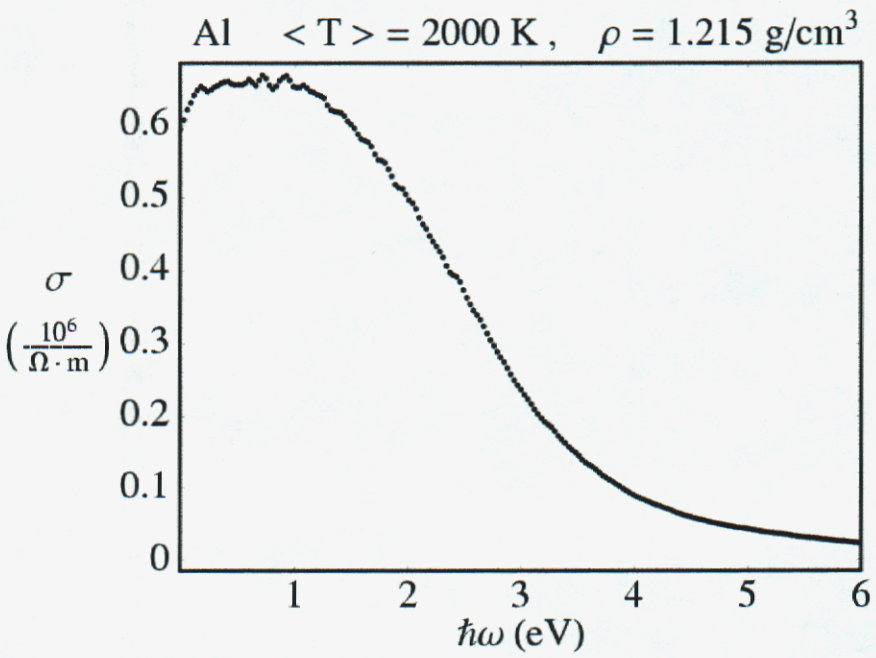
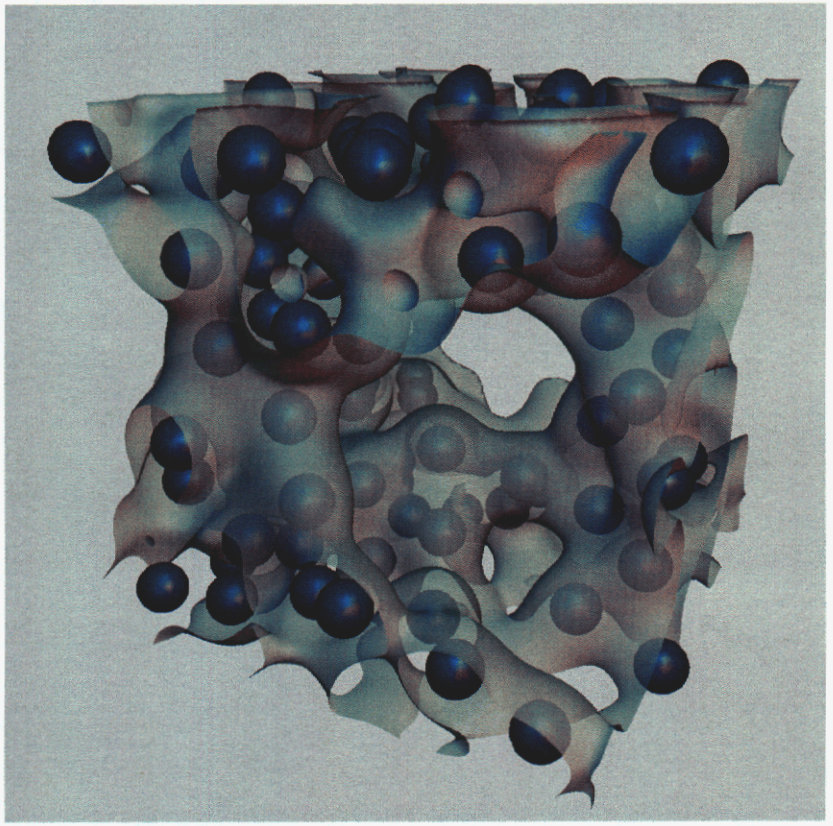
As the density drops, the optical conductivity departs from the simple Drude model behavior



A factor of two drop in density has resulted in a factor of six drop in the dc conductivity



A hint of liquid/vapor phase separation is apparent in the density plot

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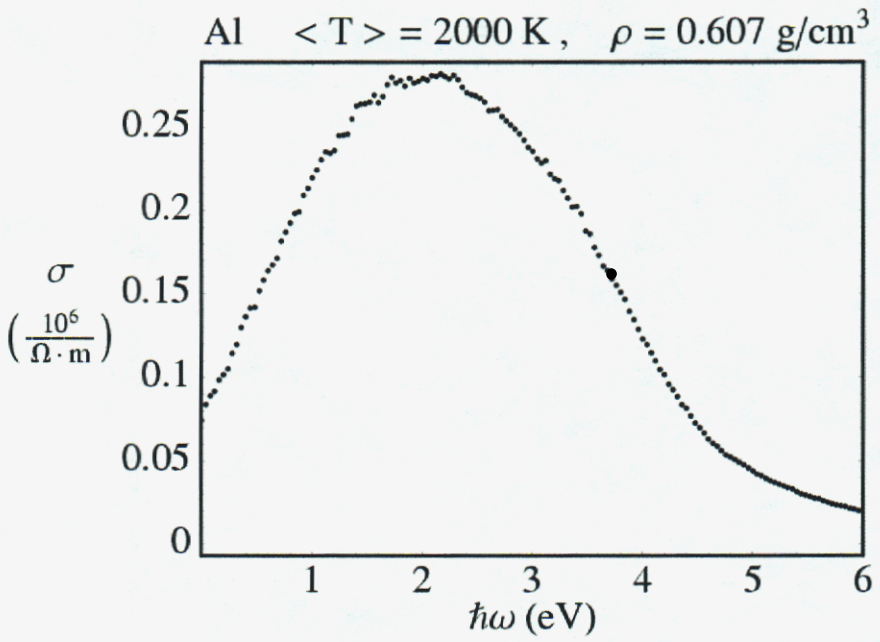
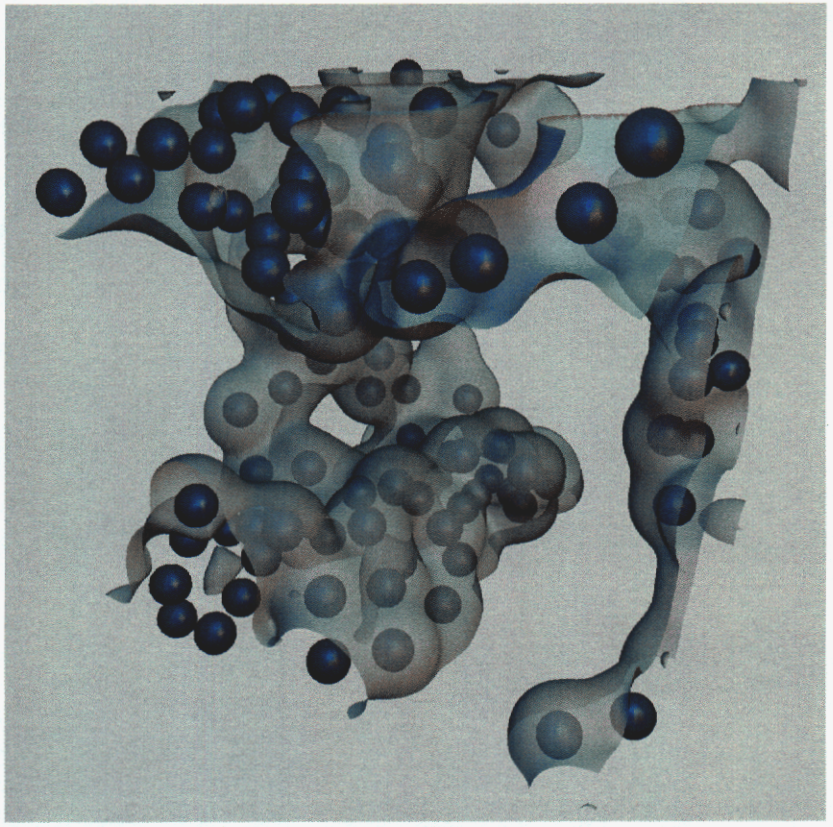
At still lower density, where phase separation is pronounced, a gap begins to form at low energy





Note the pronounced separation into liquid and void (vapor) regions

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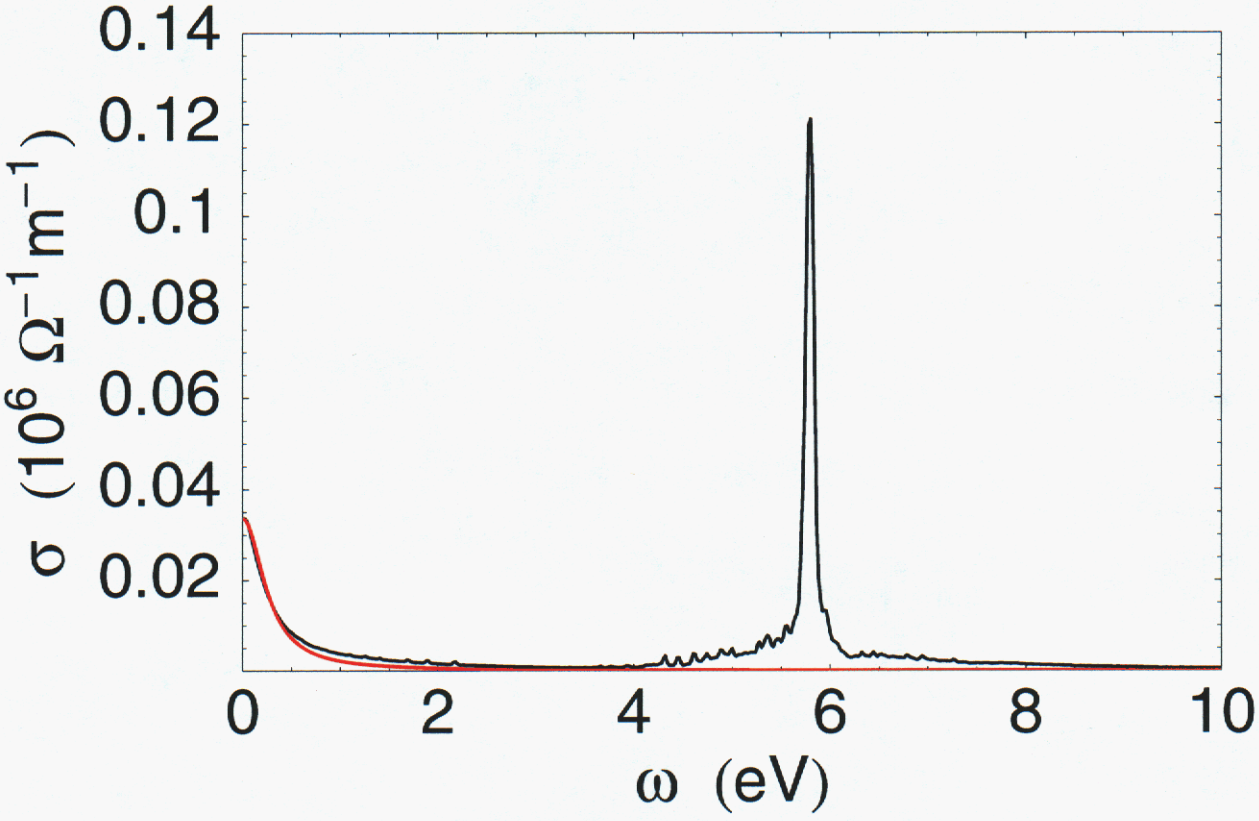
At low densities and higher temperatures, a Drude component reemerges in the optical conductivity

The optical conductivity for 0.025 g/cc at 30000 K



The peak at about 5.8 eV is characteristic of a 3s->3p transition of the isolated atom.

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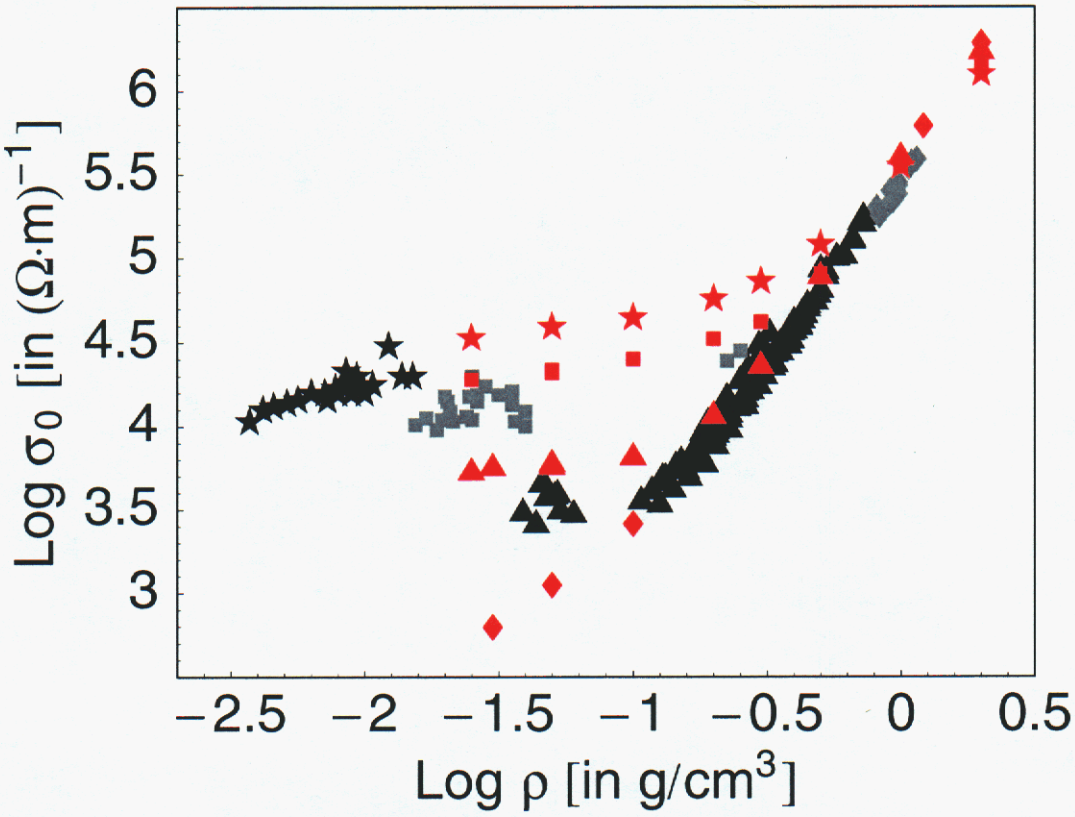
The MD-KG results are in good agreement with DeSilva's data over a two decade range of density



DeSilva and Katsouros data in black or grey, MD-KG results in red

★ 30000 K, ■ 20000 K, ▲ 10000 K, ◆ 6000 K

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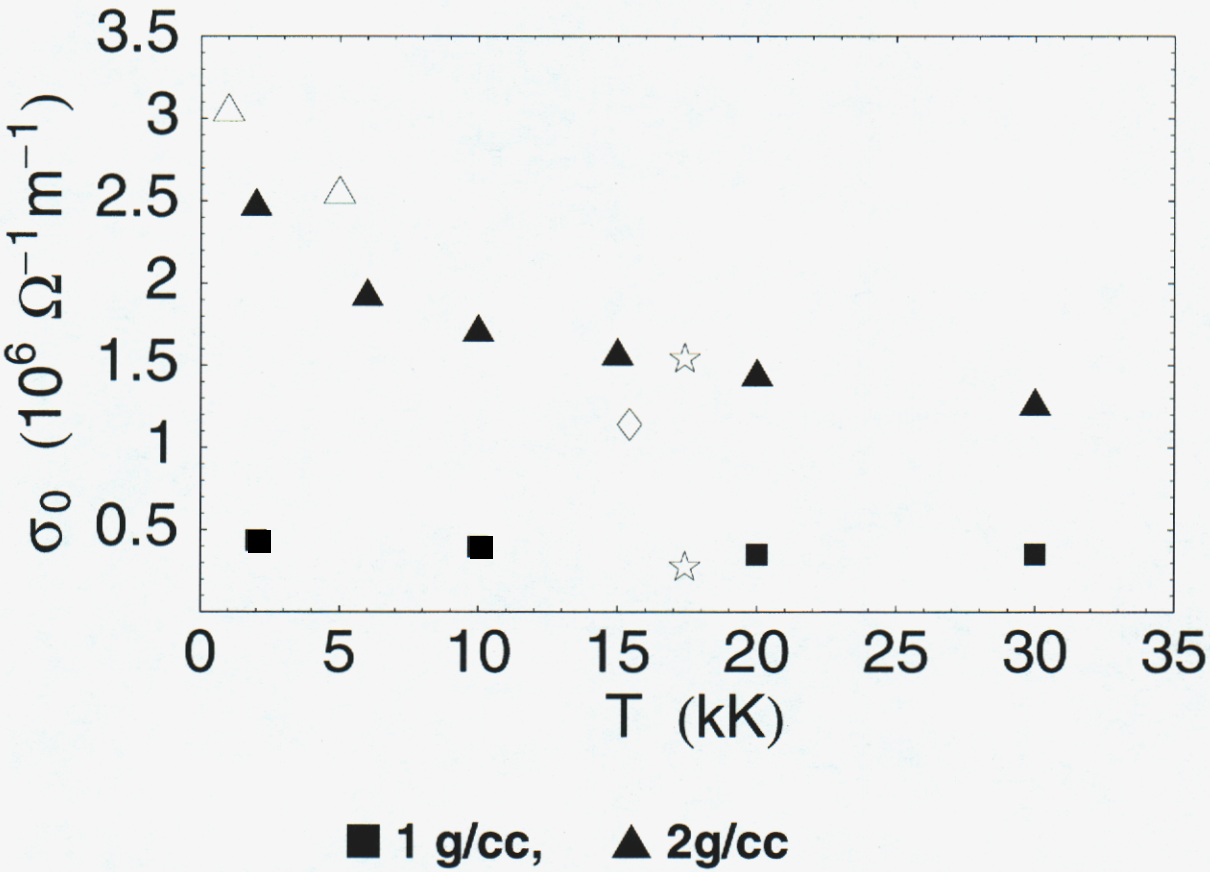
The scaling of σ_0 with temperature changes qualitatively at about 1 g/cc



Also show are similar MD-KG results from Silvestrelli (open triangles), data point from Benage, Shanahan, and Murillo (open diamond), and calculation results from Perrot and Dharma-wardana (open stars).

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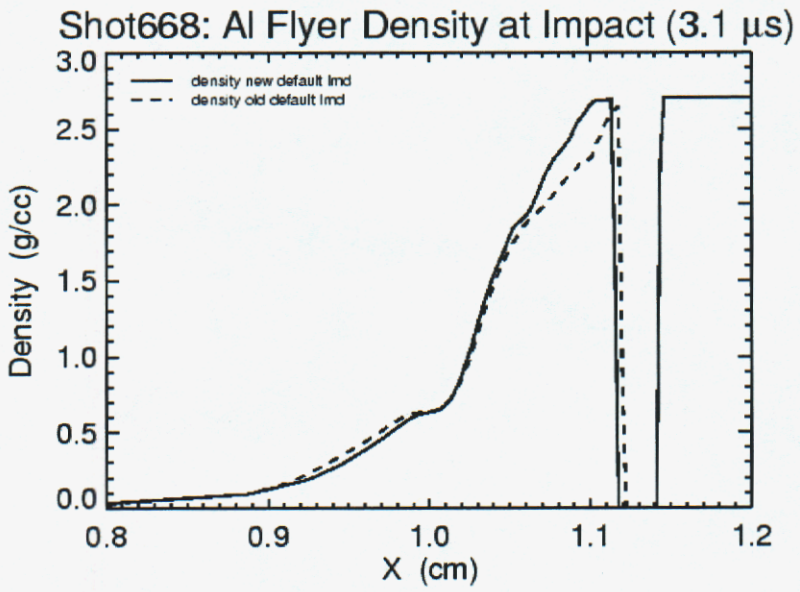
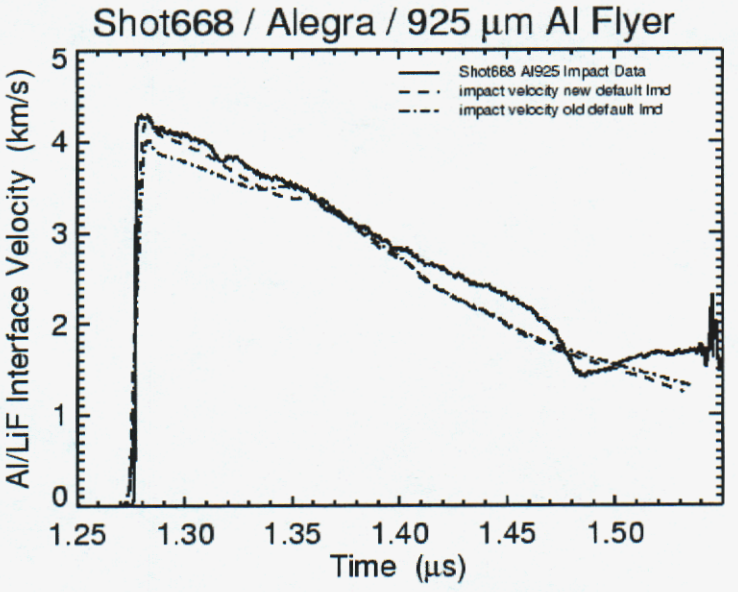
The revised conductivities result in higher fidelity simulations of magnetically launched flyer plates





ALEGRA simulation results courtesy of Ray Lemke

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One of our goals is to develop consistent EOS and transport models

Typically the equation of state, transport model, and opacity model are derived from separate formalisms and these three physics packages are generally not consistent. For example, the implicit mean ionization state of the atoms $\bar{Z}$ is often entirely different in the different models, particularly in regions of rapid changes such as the metal-insulator and liquid-vapor transitions.

The quantum molecular dynamics simulations with the associated conductivity calculations provide us with the opportunity to develop manifestly consistent equation of state and conductivity models, and also obtain, from the optical conductivity, information about the low energy optical properties of the system.

Summary

- Simple modifications to the Lee-More algorithm give substantial improvement in the accuracy of the conductivities near the metal-insulator transition
- Good agreement with the data of DeSilva and Katsouros (U. of Maryland) and Benage, Shanahan, and Murillo (LANL) is obtained
- Careful analysis of exploding wire simulations and magnetically launched flyer plates indicated probable conductivity errors up to 100% for liquid aluminum.
- Quantum molecular dynamics simulations (DFT) and Kubo-Greenwood calculations for the conductivity led to revised conductivities for the liquid Al regime.
- ALEGRA simulations of exploding wires and flyer plates with the new conductivities give quite good agreement with experiment.